Electronic Annex: **Supplementary Figures for Slotznick et al.** ***The Effects of Metamorphism on Iron Mineralogy and the Iron Speciation Redox Proxy***

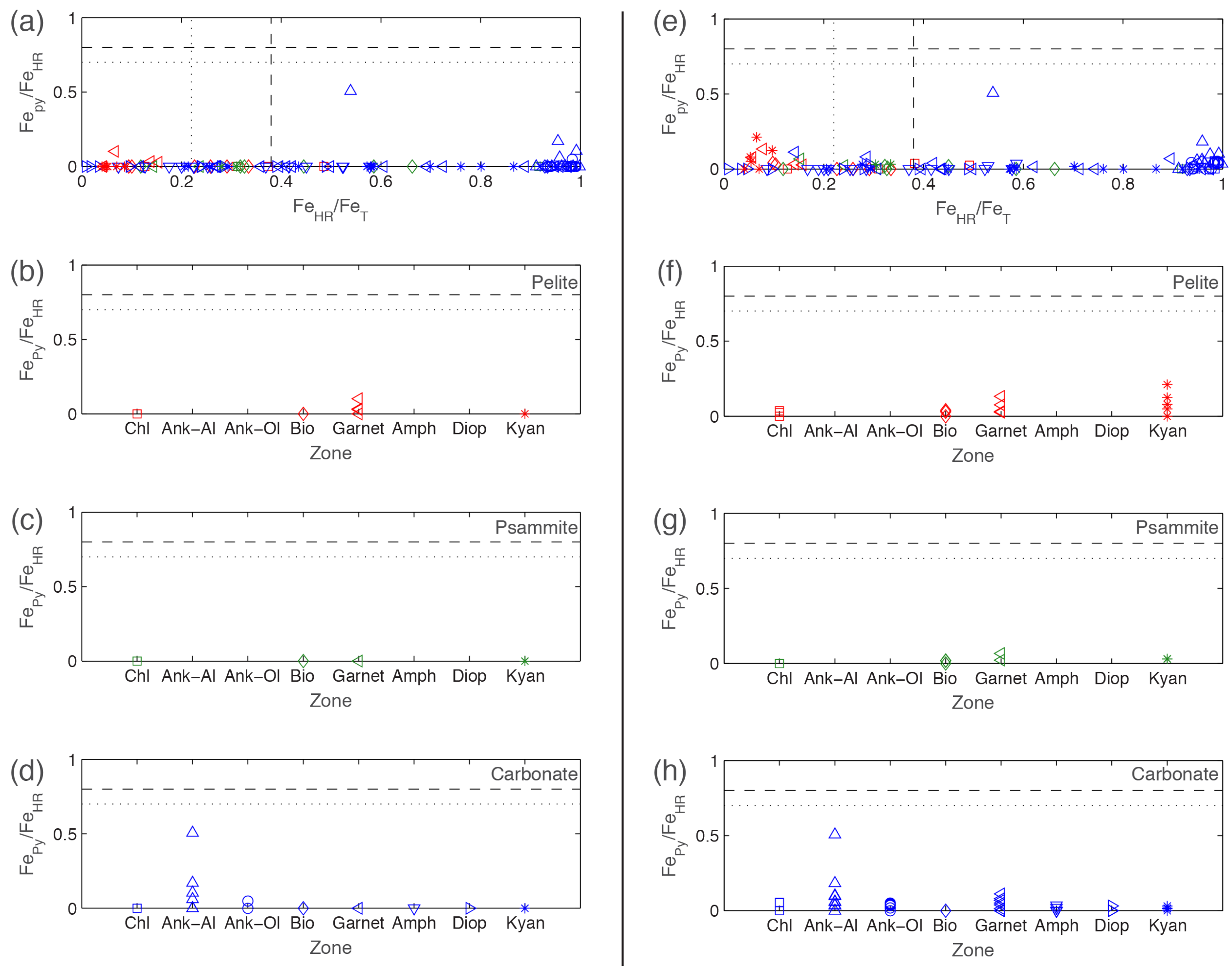

Figure EA-1: Waits River and Gile Mountain Formation data plotted in iron speciation space assuming pyrrhotite in the iron carbonate (Fe_{carb}) pool with details of Fe_{py}/Fe_{HR} ratios separated by lithology and zone. Same symbols as Figure 3; here labeled on the plots with different symbols representing the 8 different metamorphic zones color-coded by lithology. (a) Without trace minerals included. (b) With trace minerals included. Abbreviations for zones are: Chl = Chlorite, Ank-Al = Ankerite–Albite, Ank-Ol = Ankerite-Oligoclase, Bio = Biotite, Amph = Amphibole, Diop = Diopside, Kyan = Kyanite.

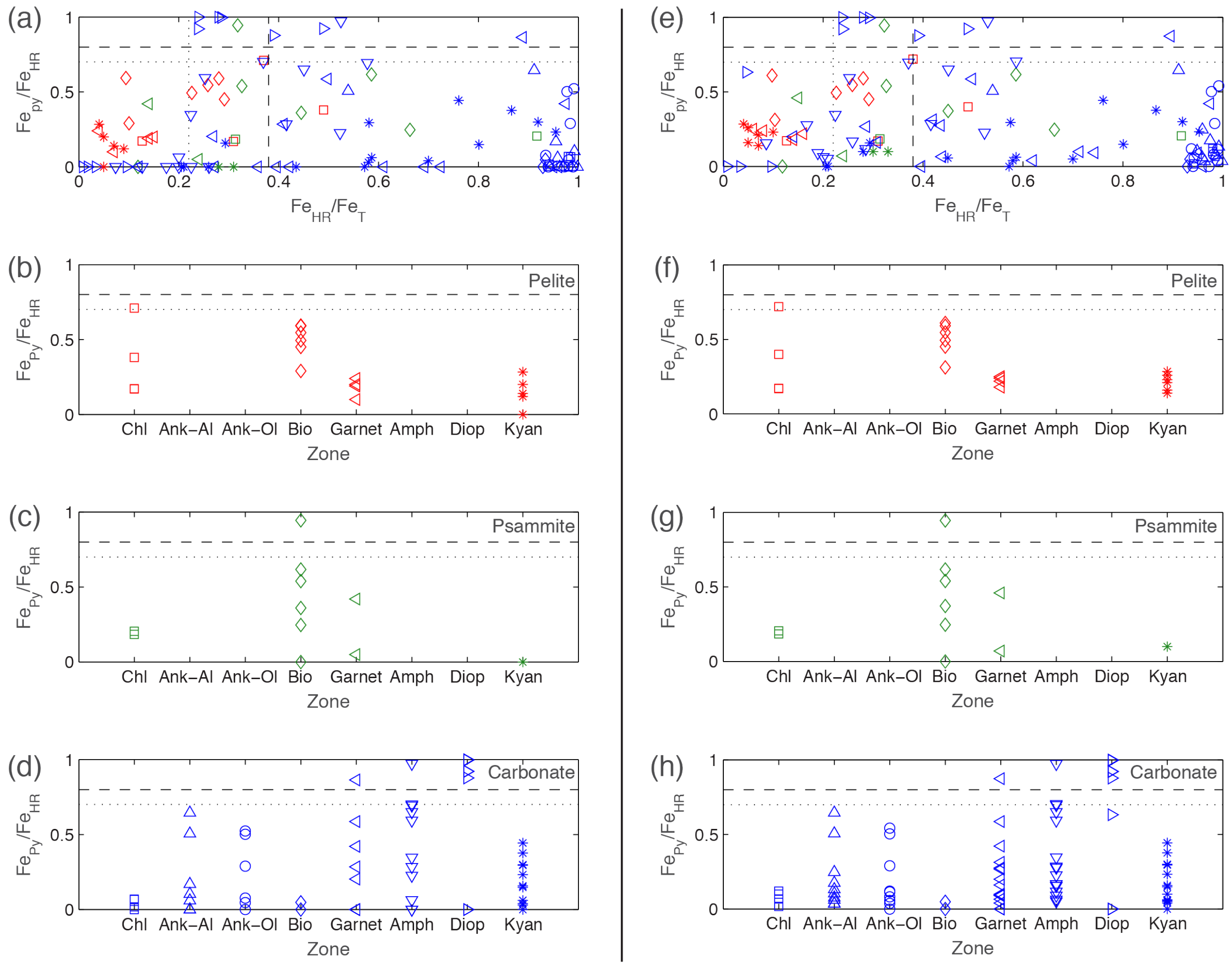

Figure EA-2: Waits River and Gile Mountain Formation data plotted in iron speciation space assuming pyrrhotite in the pyrite (Fe_{py}) pool with details of Fe_{py}/Fe_{HR} ratios separated by lithology and zone. Same symbols as Figure 3; here labeled on the plots with different symbols representing the 8 different metamorphic zones color-coded by lithology. (a) Without trace minerals included. (b) With trace minerals included. Abbreviations for zones are: Chl = Chlorite, Ank-Al = Ankerite–Albite, Ank-Ol = Ankerite-Oligoclase, Bio = Biotite, Amph = Amphibole, Diop = Diopside, Kyan = Kyanite.

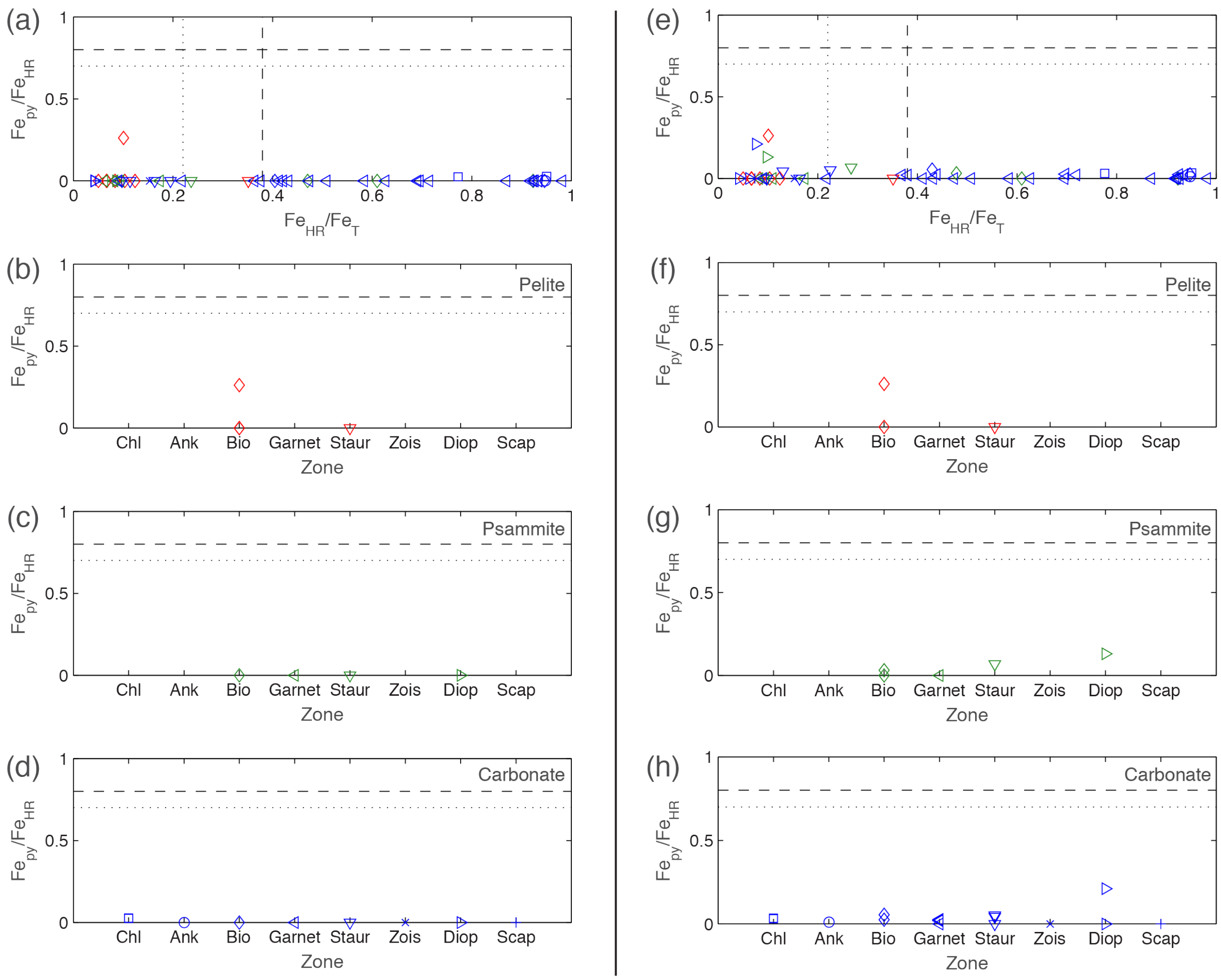

Figure EA-3: Waterville and Sangerville-Vassalboro Formation data plotted in iron speciation space assuming pyrrhotite in the iron carbonate (Fe_{carb}) pool with details of Fe_{py}/Fe_{HR} ratios separated by lithology and zone. Same symbols as Figure 6; here labeled on the plots with different symbols representing the 8 different metamorphic zones color-coded by lithology. (a) Without trace minerals included. (b) With trace minerals included. Abbreviations for zones are: Chl = Chlorite, Ank = Ankerite, Bio = Biotite, Staur = Staurolite (Amphibole), Zois = Zoisite, Diop = Diopside, Scap = Scapolite.

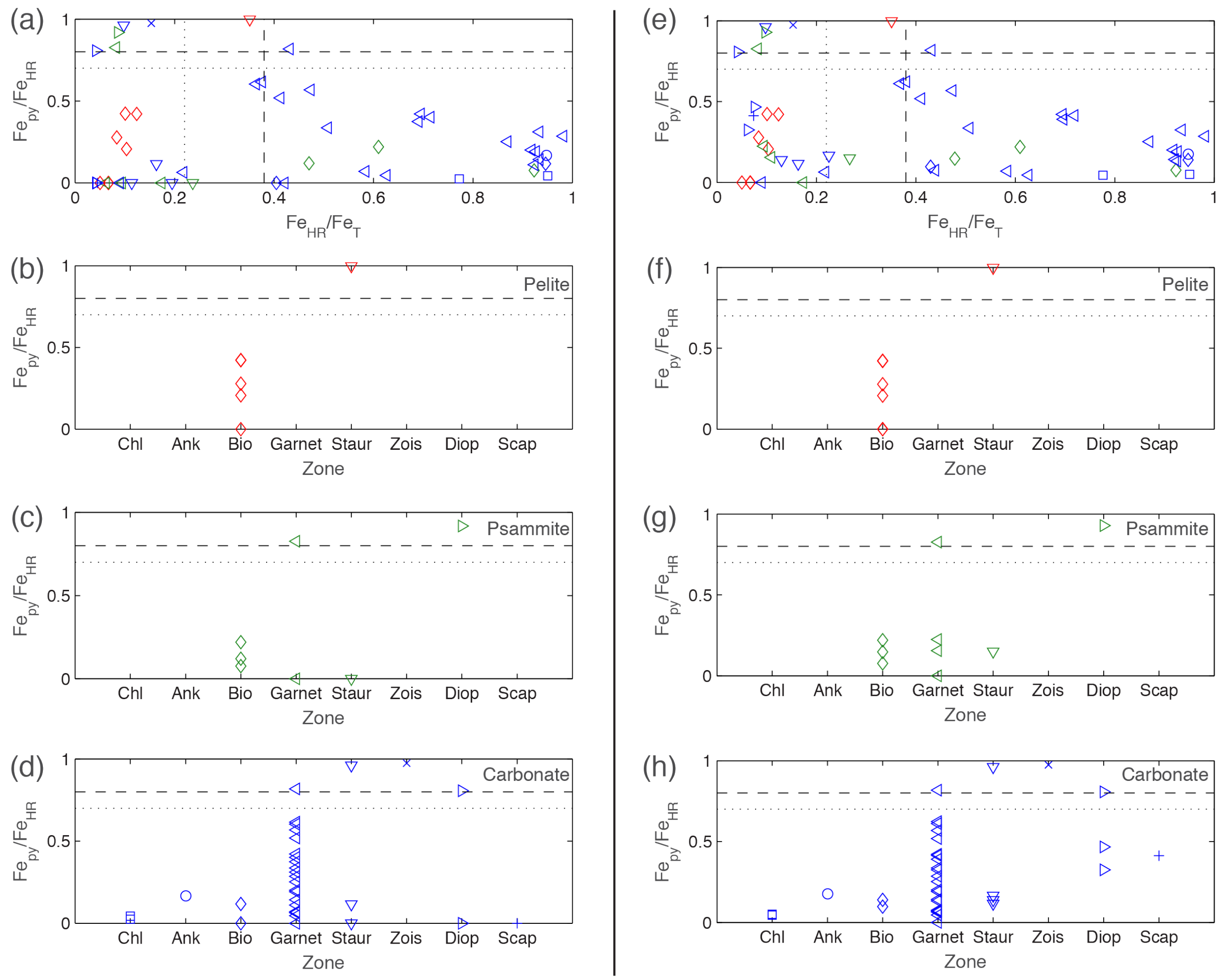

Figure EA-4: Waterville and Sangerville-Vassalboro Formation data plotted in iron speciation space assuming pyrrhotite in the pyrite (Fe_{py}) pool with details of Fe_{py}/Fe_{HR} ratios separated by lithology and zone. Same symbols as Figure 6; here labeled on the plots with different symbols representing the 8 different metamorphic zones color-coded by lithology. (a) Without trace minerals included. (b) With trace minerals included. Abbreviations for zones are: Chl = Chlorite, Ank = Ankerite, Bio = Biotite, Staur = Staurolite (Amphibole), Zois = Zoisite, Diop = Diopside, Scap = Scapolite.

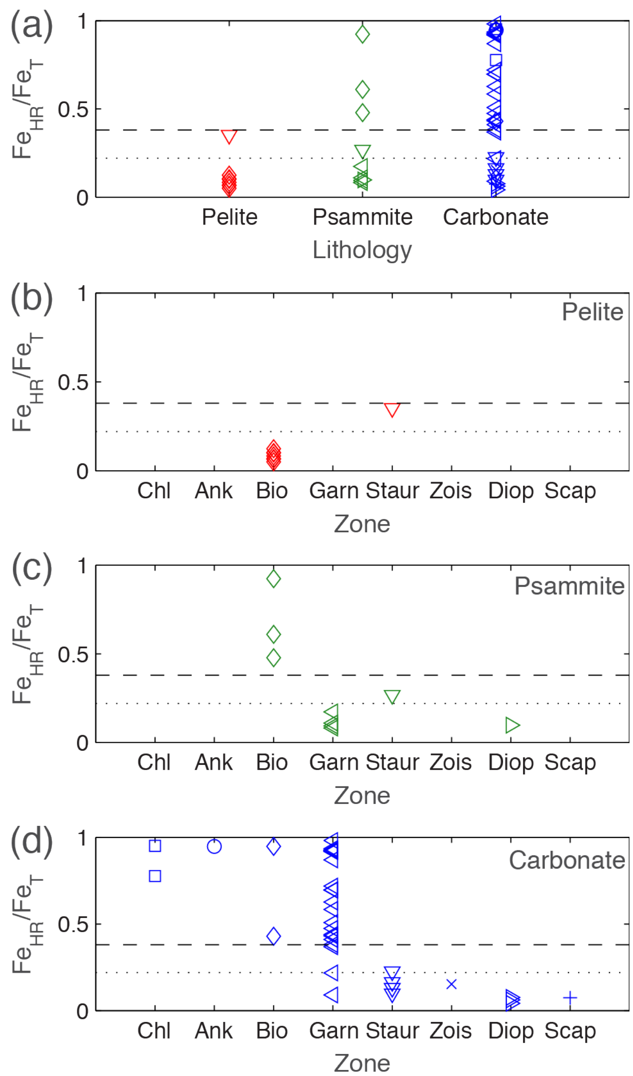

Figure EA-5: Highly reactive iron to total iron (Fe_{HR}/Fe_T) ratios in detail separated by lithology and zone for the Waterville and Sangerville-Vassalboro Formations. Same symbols as Figure 6; here labeled on the plots with symbols for metamorphic zone color-coded by lithology. Abbreviations for zones are: Chl = Chlorite, Ank = Ankerite, Bio = Biotite, Garn = Garnet, Staur = Staurolite (Amphibole), Zois = Zoisite, Diop = Diopside, Scap = Scapolite.

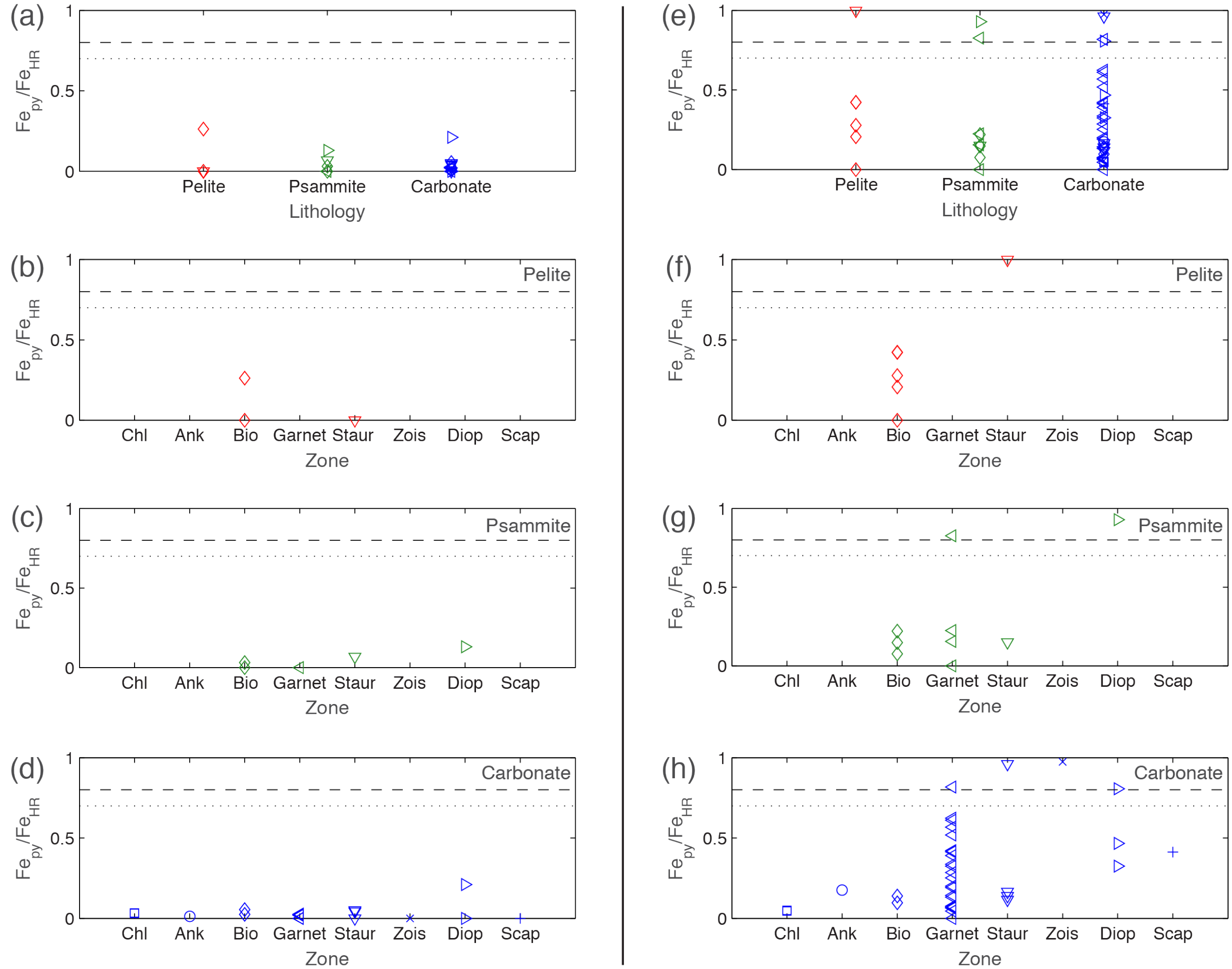

Figure EA-6: Pyrite to highly reactive iron (Fe_{py}/Fe_{HR}) ratios in detail separated by lithology and zone for the Waterville and Sangerville-Vassalboro Formations. (a-d) Pyrrhotite in the iron carbonate pool (Fe_{carb}). (e-h) Pyrrhotite in the pyrite pool (Fe_{py}). Same symbols as Figure 6; here labeled on the plots with symbols for metamorphic zone color-coded by lithology. Abbreviations for zones are: Chl = Chlorite, Ank = Ankerite, Bio = Biotite, Staur = Staurolite (Amphibole), Zois = Zoisite, Diop = Diopside, Scap = Scapolite.

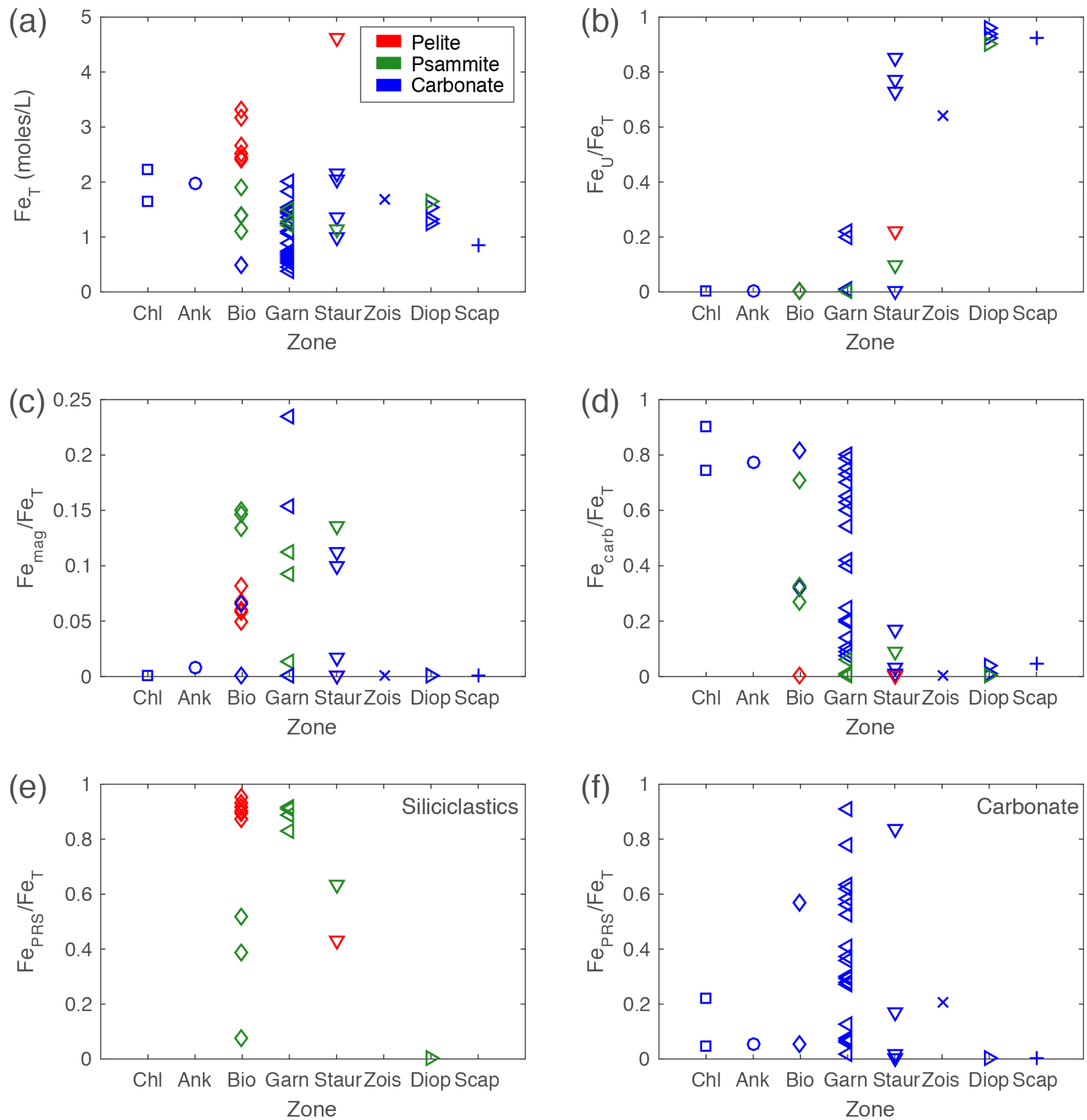

Figure EA-7: Different iron pools separated by metamorphic grade in samples from Waterville and Sangerville-Vassalboro Formations. (a) Total iron (Fe_T). (b) Ratio of unreactive silicate iron to total iron (Fe_U/Fe_T). (c) Ratio of magnetite iron pool (entirely ilmenite here) to total iron (Fe_{mag}/Fe_T). Note this is at a different scale than (b-f). (d) Ratio of carbonate iron to total iron (Fe_{carb}/Fe_T). (e) Ratio of poorly reactive sheet silicates to total iron (Fe_{PRS}/Fe_T) for pelites and psammites. (f) Ratio of poorly reactive sheet silicates to total iron (Fe_{PRS}/Fe_T) for carbonate. Same symbols as Figure 6; lithology color codes in legend in (a) with symbols for metamorphic zone labeled on plots. Abbreviations for zones are: Chl = Chlorite, Ank = Ankerite, Bio = Biotite, Garn = Garnet, Staur = Staurolite (Amphibole), Zois = Zoisite, Diop = Diopside, Scap = Scapolite. All data here is assuming pyrrhotite is in the pyrite pool (Fe_{py}).

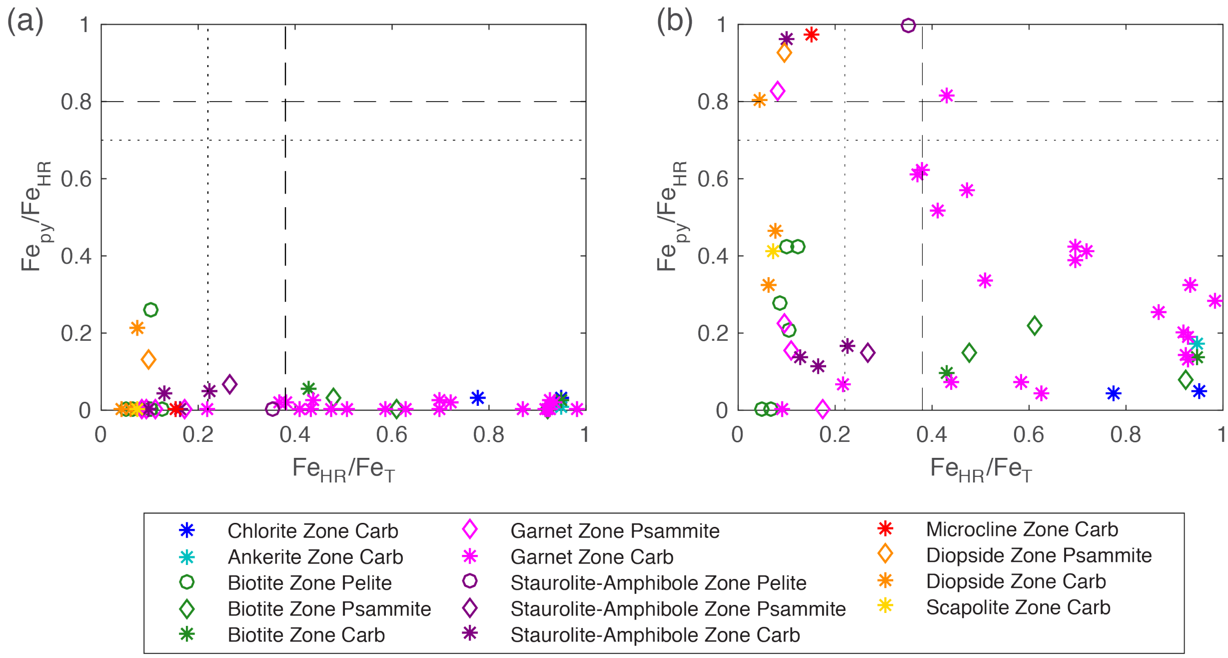

Figure EA-8: Waterville and Sangerville-Vassalboro Formations data plotted in iron speciation space. Legend is the same for both plots with different symbols representing the 3 different lithologies color-coded by metamorphic zones. Staurolite (Amphibole) stands for the pelitic schist and carbonate facies respectively. (a) Pyrrhotite in the Fe_{carb} pool. (b) Pyrrhotite in the pyrite pool (Fe_{py}).

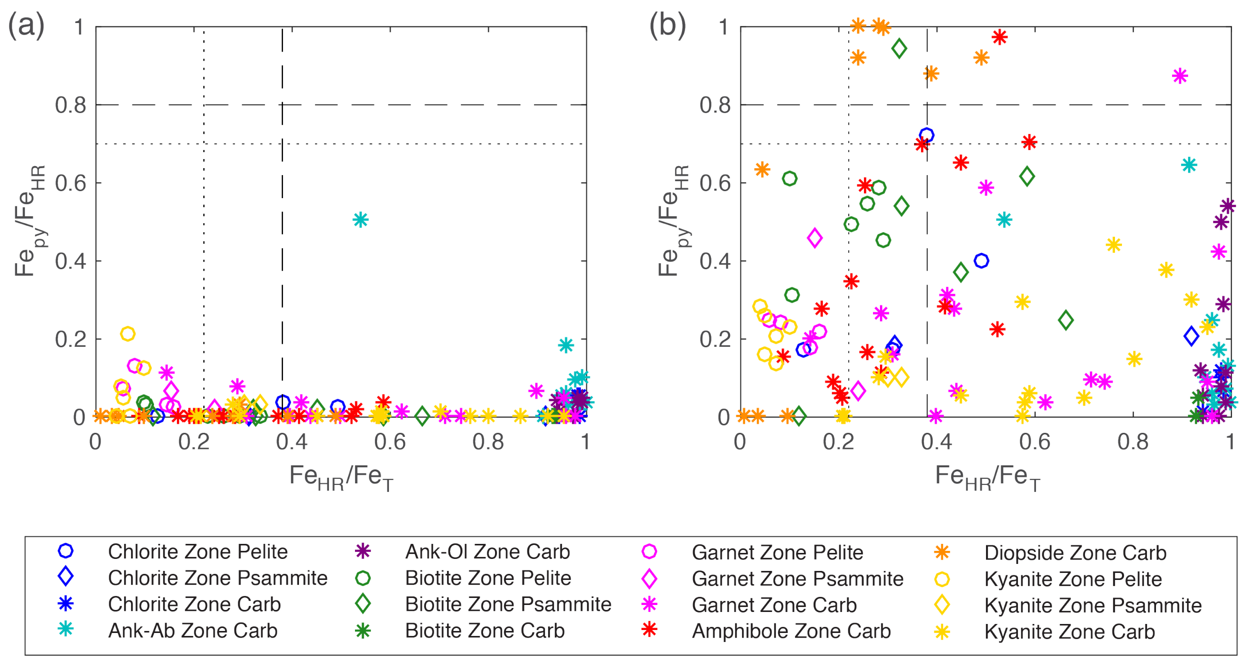

Figure EA-9: Waits River and Gile Mountain Formations data plotted in iron speciation space. Legend is the same for both plots with different symbols representing the 3 different lithologies color-coded by metamorphic zones. Ank-Al stands for Ankerite-Albite and Ank-Ol stands for Ankerite-Oligoclase. (a) Pyrrhotite in the iron carbonate (Fe_{carb}) pool. (b) Pyrrhotite in the pyrite pool (Fe_{py}). Fe_{py}/Fe_{HR} is the ratio of pyrite to highly reactive iron and Fe_{HR}/Fe_T is the ratio of highly reactive to total iron.

Table EA-1: Student T-test results comparing lithological differences of iron speciation ratios

Ratio	Pop. A	Pop. B	Pyrrhotite Pool	p value	Hypothesis Test (alpha =0.05)
Fe_{HR}/Fe_T	Carbonate	Psammite	N/A	0.0350526	Significantly Different
Fe_{HR}/Fe_T	Carbonate	Pelite	N/A	9.172529e-08	Significantly Different
Fe_{HR}/Fe_T	Pelite	Psammite	N/A	0.0014041	Significantly Different
Fe_{py}/Fe_{HR}	Carbonate	Psammite	Carbonate	0.6391808	Not Significantly Different
Fe_{py}/Fe_{HR}	Carbonate	Pelite	Carbonate	0.2272398	Not Significantly Different
Fe_{py}/Fe_{HR}	Pelite	Psammite	Carbonate	0.1196756	Not Significantly Different
Fe_{py}/Fe_{HR}	Carbonate	Psammite	Pyrite	0.5452026	Not Significantly Different
Fe_{py}/Fe_{HR}	Carbonate	Pelite	Pyrite	0.3340458	Not Significantly Different
Fe_{py}/Fe_{HR}	Pelite	Psammite	Pyrite	0.8829638	Not Significantly Different

Analysis performed using Matlab

Table EA-2: Mann-Kendall Test for non-parametric trend analysis of iron speciation ratio variation across metamorphic grade

Ratio	Lithology	Pyrrhotite Pool	Tau	S Statistic	p value	Hypothesis Test (alpha =0.05)
Fe_{HR}/Fe_T	All	N/A	-0.3174973	-2045	4.600200e-07	Significant Decreasing Trend
Fe_{HR}/Fe_T	Carbonate	N/A	-0.4314965	-1433	6.854410e-09	Significant Decreasing Trend
Fe_{HR}/Fe_T	Psammite	N/A	-0.2424242	-16	2.666298e-01	Not Significant Decreasing Trend
Fe_{HR}/Fe_T	Pelite	N/A	-0.6210526	-118	9.997816e-05	Significant Decreasing Trend
Fe_{py}/Fe_{HR}	Carbonate	Carbonate	-0.3038241	-1009	2.231608e-06	Significant Decreasing Trend
Fe_{py}/Fe_{HR}	Psammite	Carbonate	0.5454545	36	5.519839e-03	Significant Increasing Trend
Fe_{py}/Fe_{HR}	Pelite	Carbonate	0.3157895	60	4.225611e-02	Significant Increasing Trend
Fe_{py}/Fe_{HR}	Carbonate	Pyrite	0.1035833	344	1.646086e-01	Significant Increasing Trend
Fe_{py}/Fe_{HR}	Psammite	Pyrite	-0.1212121	-8	6.034409e-01	Not Significant Decreasing Trend
Fe_{py}/Fe_{HR}	Pelite	Pyrite	-0.3157895	-60	4.976841e-02	Significant Decreasing Trend

Analysis performed in Matlab using code from: Burkey, Jeff. May 2006. A non-parametric monotonic trend test computing Mann-Kendall Tau, Tau-b, and Senís Slope written in Mathworks-MATLAB implemented using matrix rotations. King County, Department of Natural Resources and Parks, Science and Technical Services section. Seattle, Washington. USA. http://www.mathworks.com/matlabcentral/fileexchange/authors/23983